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## Roles of adhesives in forming mechanically robust superhydrophobic coatings

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Superhydrophobic coatings, renowned for their remarkable water repellence, have attracted considerable attention for diverse applications, but their practical application is often hindered by insufficient mechanical robustness. Among various strategies proposed to address this limitation, the use of adhesives has emerged as a highly promising approach, offering advantages such as low cost, facile processing, and compatibility with diverse substrates. This review systematically summarizes the enhancement mechanisms, recent research progress, commercialization status, and common testing methods for the mechanical robustness of mechanically robust superhydrophobic coatings based on adhesives. First, the fundamental mechanisms by which adhesives enhance mechanical robustness, including interfacial adhesion enhancement, structural protection, and functional augmentation, are thoroughly analyzed. Next, recent advances in mechanically robust superhydrophobic coatings based on different types of adhesives, are reviewed. Subsequently, this review summarizes recent developments in mechanically robust superhydrophobic coatings prepared via different adhesive application methods. Additionally, the review summarizes the commercial products currently available, analyzing mechanical robustness and highlighting the most performant products. The common testing methods for the mechanical robustness of superhydrophobic coatings are also introduced. Finally, we discuss the challenges faced in this field and outline potential research directions to address the current limitations. This review aims to provide valuable guidance for the rational design of high-performance mechanically robust superhydrophobic coatings based on adhesives.

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### 1. Introduction

Although the phenomenon of superhydrophobicity had been first documented by Ollivier in 1907,<sup>1</sup> it remained largely underexplored for much of the 20th century. Interest in the field was reignited in 1997 when Barthlott and Neinhuis systematically revealed the self-cleaning mechanism of lotus leaves.<sup>2</sup> Since then, superhydrophobic coatings have garnered

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significant attention across both academic and industrial sectors.<sup>3–9</sup> According to the Web of Science database, the number of articles and patents related to “superhydrophobic\*”

has grown exponentially since 2000, surpassing 27 000 and 4300 by December 2024, respectively (Fig. 1a and b).

The fundamental principle underlying superhydrophobic coatings is the creation of surface structures that combine high

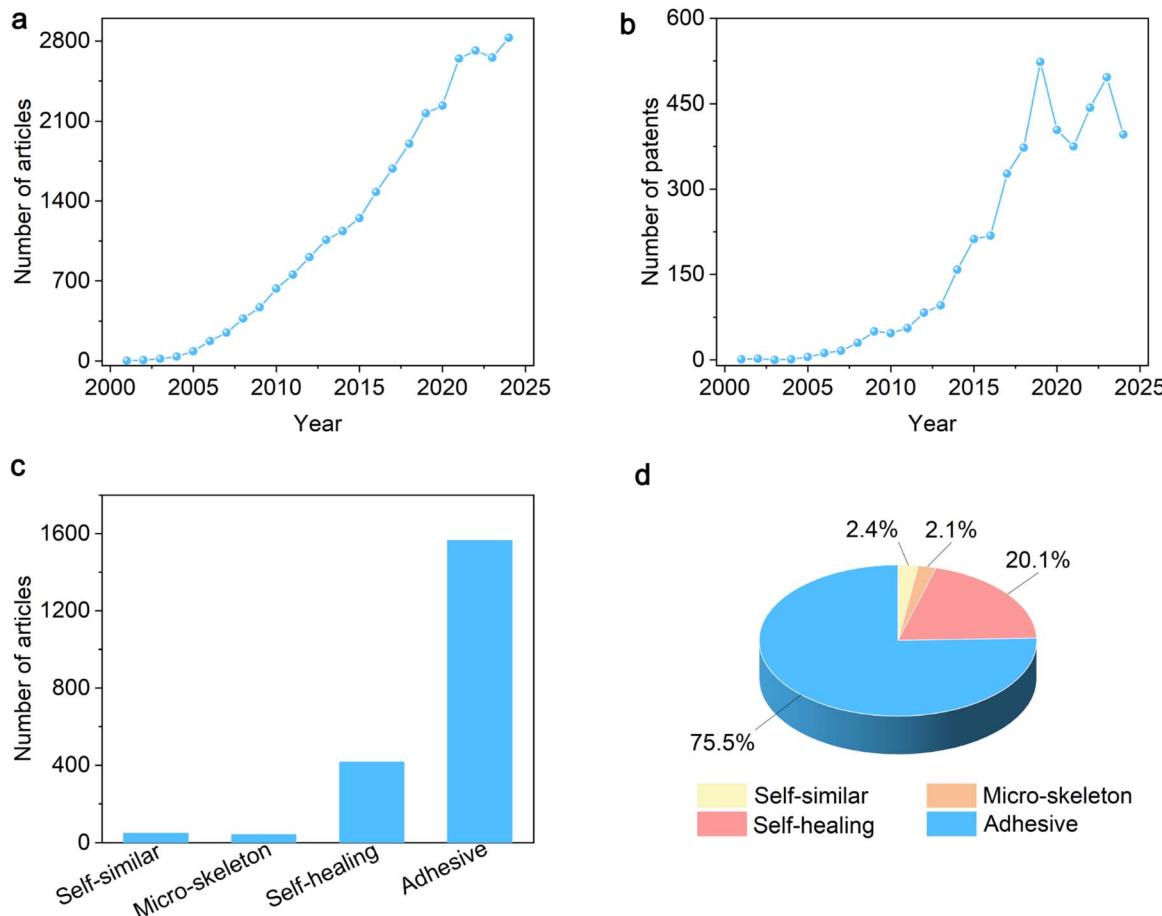


Fig. 1 Number of (a) articles and (b) patents related to superhydrophobic coatings from 2001 to 2024. (c) Number and (d) proportion of articles utilizing different strategies to enhance the mechanical robustness of superhydrophobic coatings as of December 2024.



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roughness with low surface energy, leading to water droplets exhibiting contact angles greater than 150° and sliding angles less than 10°.<sup>10,11</sup> Building on this principle, researchers have developed a variety of fabrication methods, including etching,<sup>12–16</sup> templating,<sup>17–19</sup> electrospinning,<sup>20–22</sup> chemical vapor deposition,<sup>23–25</sup> dip-coating,<sup>26–28</sup> and spray-coating.<sup>29–31</sup> Due to their exceptional wetting behavior, superhydrophobic coatings have shown great promise in diverse applications, such as self-cleaning surfaces,<sup>32–34</sup> oil-water separation,<sup>35–37</sup> anti-corrosion,<sup>38–40</sup> anti-icing, and so on.<sup>41–43</sup>

Despite their impressive performance in laboratory settings, superhydrophobic coatings encounter a significant challenge when it comes to practical applications: limited mechanical robustness.<sup>44–46</sup> The micro-/nanostructures that are essential to their superhydrophobicity are inherently fragile and highly susceptible to damage from external forces, such as abrasion and impact.<sup>47–50</sup> When these micro-/nanostructures are compromised or when low-surface-energy nanoparticles detach, the coating's superhydrophobicity can degrade substantially, or even be completely lost.<sup>51,52</sup> In response to this challenge, numerous strategies have been proposed over the past decade to enhance the mechanical robustness of superhydrophobic coatings.<sup>53–55</sup> Typical strategies include imparting self-healing capabilities to the coatings,<sup>45,56,57</sup> constructing self-similar structures,<sup>58–60</sup> and incorporating hard micro-skeletons to protect the nanostructures.<sup>58,61–63</sup> While these methods have led to significant advancements, they often face practical limitations such as high production costs, complex fabrication processes, and difficulties in scaling up for industrial applications, all of which hinder their widespread use.

In addition to the strategies mentioned above, the use of adhesives to enhance the mechanical robustness of superhydrophobic coatings has emerged as one of the most promising and actively researched approaches, owing to their low cost, compatibility with various substrates, and scalability for large-area applications.<sup>64–66</sup> According to the Web of Science database, over 1500 articles have reported the mechanically robust superhydrophobic coatings based on adhesives by December 2024, accounting for approximately 75.5% of all studies focused on enhancing the mechanical robustness of superhydrophobic coatings (Fig. 1c and d). Nevertheless, current mechanically robust superhydrophobic coatings based on adhesives still face several challenges, including the limited improvement in mechanical robustness, poor superhydrophobicity (particularly under dynamic or real-world conditions) and weather resistance.<sup>67–70</sup> In addition, the widespread use of volatile organic compounds (VOCs) in many fabrication processes raises environmental and safety concerns.<sup>71,72</sup> Moreover, achieving consistent adhesion strength across a wide range of substrates remains difficult, especially on hydrophobic surfaces.<sup>73,74</sup>

To date, numerous review articles have reviewed the principles and fabrication methods of superhydrophobic coatings and research progress of mechanically robust superhydrophobic coatings, as well as their mechanical robustness testing methods.<sup>3,44,53–55,75–79</sup> Although some reviews on mechanically robust superhydrophobic coatings have briefly

mentioned the progress of mechanically robust superhydrophobic coatings based on adhesives,<sup>53,54</sup> a comprehensive and systematic review focusing on the enhancement mechanisms, adhesive application methods, and commercial status of the mechanically robust superhydrophobic coatings based on adhesives is still lacking, yet it is crucial for advancing their development.

Therefore, to facilitate the further research and application of mechanically robust superhydrophobic coatings based on adhesives, it is essential to systematically review their enhancement mechanisms, recent research progress, commercialization status, and common testing methods for mechanical robustness. This review aims to provide a comprehensive overview encompassing the following aspects (Fig. 2): (1) a detailed analysis of the microscopic mechanisms responsible for the improved mechanical robustness imparted by adhesives; (2) recent advances in mechanically robust superhydrophobic coatings based on different types of adhesives; (3) recent developments in mechanically robust superhydrophobic coatings based on adhesives *via* different adhesive application methods; (4) a summary of commercialized mechanically robust superhydrophobic coatings based on adhesives worldwide; and (5) an introduction of common testing methods for mechanical robustness. Finally, this review highlights the key challenges and future directions for the development of high-performance mechanically robust superhydrophobic coatings based on adhesives.

## 2. Mechanisms of adhesives in enhancing the mechanical robustness of superhydrophobic coatings

Among the various types of superhydrophobic coatings, coatings formed from low-surface-energy nanoparticles are among the most widely studied and applied.<sup>60,80–83</sup> Typically, these coatings are created by modifying nanoparticles (e.g., silica, titania and attapulgite) with low-surface-energy compounds (e.g., alkylsilanes, aliphatic acids and paraffin), followed by deposition techniques like spray-coating or dip-coating.<sup>26,27,84,85</sup> However, the structural integrity of these coatings often relies predominantly on weak van der Waals forces to maintain adhesion between the nanoparticles and the substrate or among the particles themselves. Consequently, these coatings are highly vulnerable to mechanical damage from abrasion, impact, or shear forces, resulting in a rapid loss of superhydrophobicity.

In recent years, the incorporation of adhesives has emerged as a highly effective strategy to enhance the mechanical robustness of these superhydrophobic coatings.<sup>86–88</sup> The functional roles of adhesives in these systems can be broadly classified into three principal mechanisms: interfacial adhesion enhancement, structural protection, and functional augmentation.<sup>58,89–92</sup> First, adhesives improve interfacial adhesion between nanoparticles and the substrate, as well as among the nanoparticles themselves, through physical interactions (e.g., hydrogen bonding and van der Waals forces) and/or



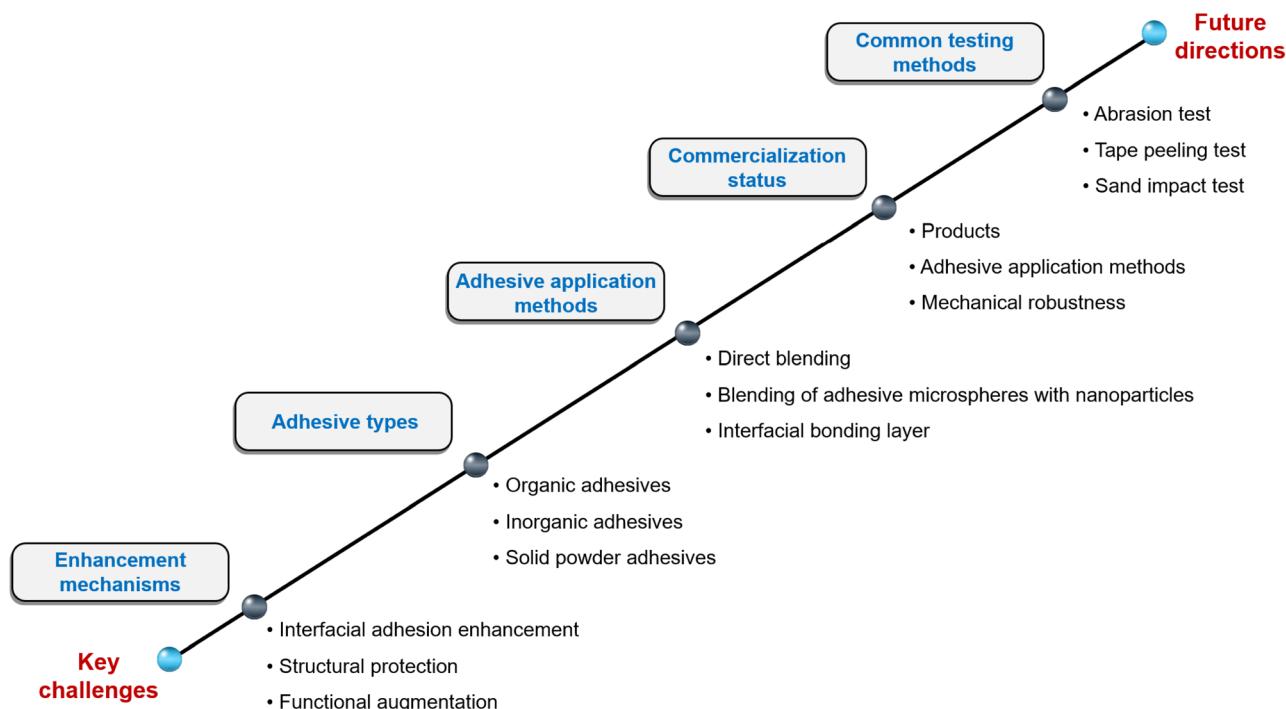


Fig. 2 Contents of this review about roles of adhesives in forming mechanically robust superhydrophobic coatings.

chemical bonding (*e.g.*, covalent and coordination bonds). Second, adhesives can form rigid microstructures that serve as “skeletons”, shielding damage to the low-surface-energy nanoparticles from external mechanical stresses. Third, certain adhesives have additional functionalities, such as self-healing capabilities or elasticity, which further contribute to the coating's mechanical robustness.

## 2.1 Interfacial adhesion enhancement

Adhesives, acting as “bridge” molecules, can effectively fill the gaps between low-surface-energy nanoparticles, significantly enhancing the van der Waals forces between the nanoparticles, as well as between the nanoparticles and the substrate.<sup>90,93,94</sup> Additionally, the hydroxyl and carboxyl groups in the adhesives can form hydrogen bonds with the hydroxyl groups on the nanoparticles or substrate surface, further strengthening these interactions.<sup>90,95</sup> Jiao *et al.* fabricated a superamphiphobic coating by spray-coating a suspension of fluorinated SiO<sub>2</sub> nanoparticles and ceramic adhesive.<sup>95</sup> The hydroxyl groups in the ceramic adhesive formed numerous hydrogen bonds with the substrate, significantly enhancing the adhesion strength between the coating and the substrate (Fig. 3a).

Beyond these physical interactions, the adhesion can be further improved through the formation of chemical bonds.<sup>90,91,96–99</sup> This process relies on the presence of reactive functional groups (*e.g.*, C=C, -SiOH and -NH<sub>2</sub>) on both the adhesives and the nanoparticles/substrates. For example, Zhao *et al.* synthesized a siloxane-modified hyperbranched waterborne polyurethane rich in Si-OH groups, which was combined with fluorinated silica nanoparticles to create a mechanically

robust superhydrophobic coating.<sup>96</sup> The abundant polar groups in siloxane-modified hyperbranched waterborne polyurethane formed Si–O–Si covalent bonds with both the substrate and the nanoparticles, resulting in a robust crosslinked network (Fig. 3b). The coating retained its superhydrophobicity after 140 cycles of sandpaper abrasion.

Furthermore, the hydroxyl groups in the adhesives can interact with metal substrates to form coordination bonds, thus enhancing the adhesion strength between the coating and the substrate.<sup>100</sup> Li *et al.* demonstrated that the hydroxyl groups in an aluminum phosphate adhesive formed numerous hydrogen bonds with the oxygen atoms on the surface of fluorinated attapulgite, while also coordinating with metal atoms on the metal substrate (Fig. 3c).<sup>100</sup> This significantly enhanced the mechanical robustness of the coating, which retained good superhydrophobicity after 200 cycles of sandpaper abrasion.

## 2.2 Structural protection

The mechanism of structural protection primarily involves the formation of rigid adhesive microstructures that act as “skeletons” to withstand mechanical stresses during abrasion, thereby effectively protecting the low-surface-energy nanostructures of superhydrophobic coatings.<sup>58,92,101</sup> The formation of the adhesive microstructures can occur through several approaches. One method involves dispersing hard micro-sized particles in the adhesive and applying this mixture onto a substrate to create a bonding layer with a microstructure. Then, low-surface-energy nanoparticles are coated onto their surface to form the mechanically robust superhydrophobic coating. For example, Zhang *et al.* constructed a bonding layer

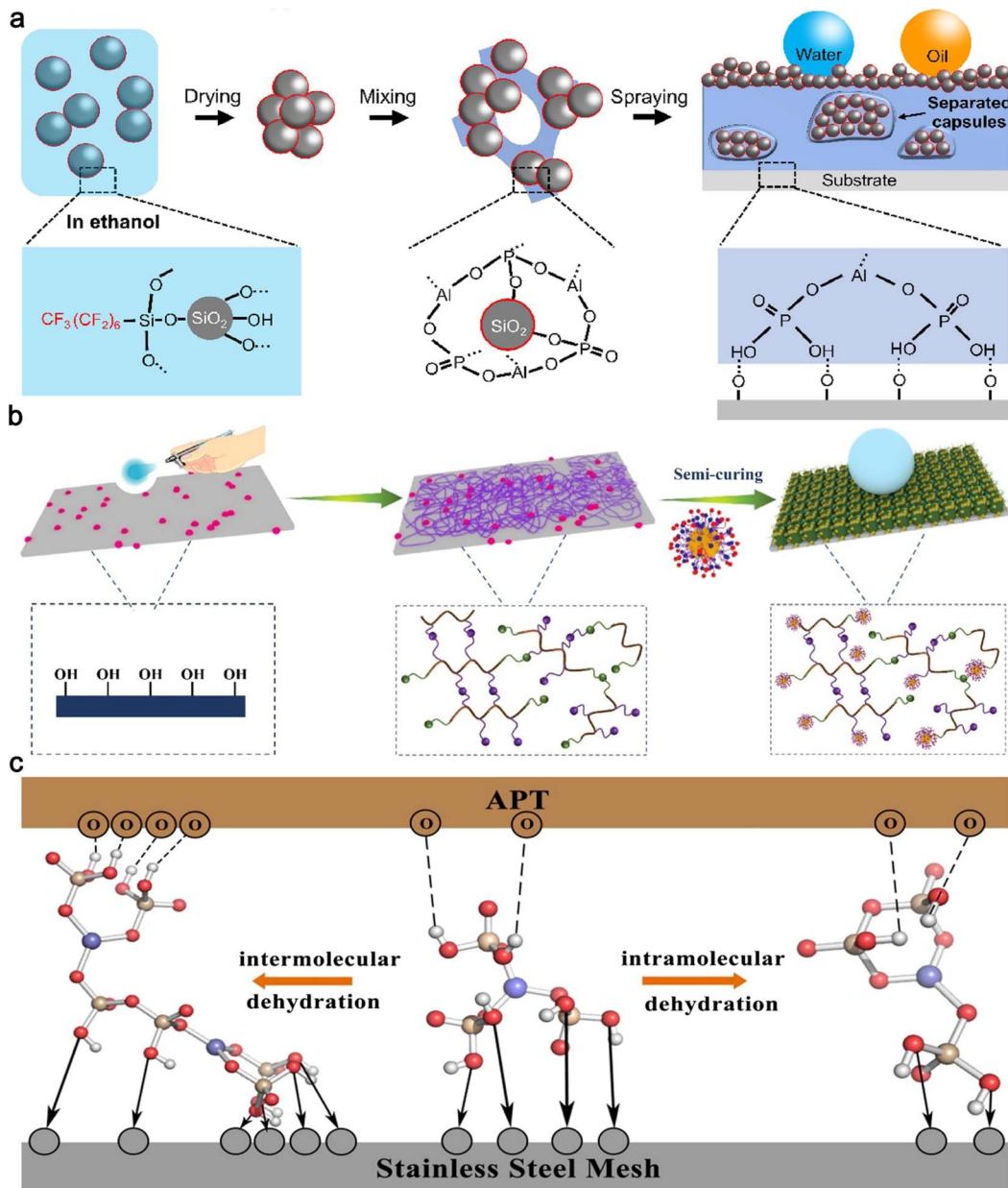


Fig. 3 Schematic illustration of the mechanism of adhesives in enhancing interfacial adhesion between nanoparticles and the substrate, as well as among the nanoparticles themselves, through formation of (a) hydrogen bonding, (b) covalent bonding, and (c) coordination bonding. (a) Reproduced with permission from ref. 95. Copyright 2022, Elsevier. (b) Reproduced with permission from ref. 96. Copyright 2022, Elsevier. (c) Reproduced with permission from ref. 100. Copyright 2019, Elsevier.

with a rigid microstructure by incorporating polypropylene microparticles into a polystyrene adhesive. Then, low-surface-energy silica nanoparticles were sprayed onto the bonding layer to create a mechanically robust superhydrophobic coating (Fig. 4a). This coating retained excellent superhydrophobicity after 150 cycles of sandpaper abrasion.<sup>92</sup>

Another way to form microstructures is by regulating the dispersion state of adhesives in solution through techniques such as non-solvent induced phase separation. Then, they are sprayed onto a substrate surface, forming rigid adhesive microstructures that effectively protect the low-surface-energy

nanoparticles.<sup>53,58</sup> Wei *et al.* induced phase separation of a silicone-modified polyester adhesive using ethanol (non-solvent) to form the composite microspheres composed of the adhesive and low-surface-energy silica nanoparticles. These microspheres were then sprayed onto an aluminum alloy surface, forming a mechanically robust superamphiphobic coating (Fig. 4b). During the spraying process, the non-solvent evaporates first, leading to partial dissolution and connection of the phase-separated adhesive microspheres, which formed a rigid microstructure that significantly enhanced the mechanical robustness of the coating.<sup>58</sup>



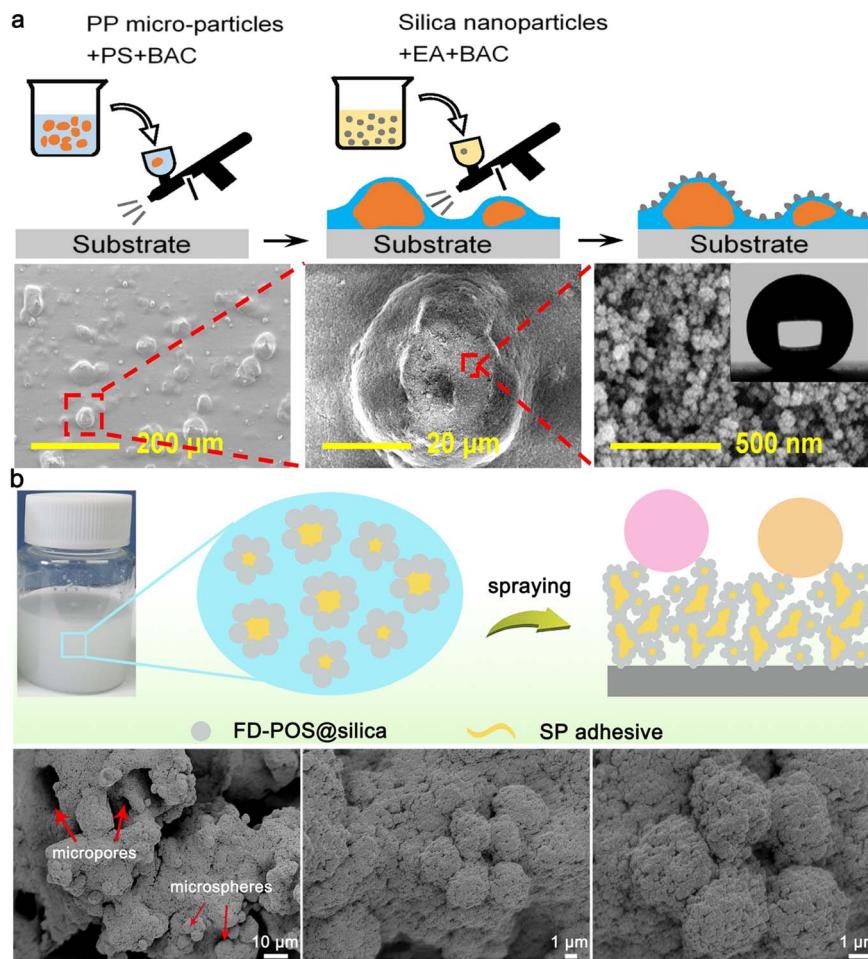


Fig. 4 Schematic illustration and scanning electron microscopy (SEM) images of the mechanically robust superhydrophobic coatings with micro-skeletons prepared (a) using a bonding layer with microstructure or (b) *via* non-solvent induced phase separation. (a) Reproduced with permission from ref. 92. Copyright 2021, Elsevier. (b) Reproduced with permission from ref. 58. Copyright 2022, WILEY-VCH.

### 2.3 Functional augmentation

Some adhesives possess self-healing properties, which, when triggered by stimuli such as heat, moisture or light, promote the migration and rearrangement of polymer chains to repair micro-damage or surface scratches. A typical example of such adhesives is shape-memory polymers. Introducing these types of adhesives into superhydrophobic coatings can endow the coatings with self-healing capabilities, enabling the repair of damaged micro-/nanostructures and significantly enhancing the mechanical robustness of the superhydrophobic coatings.<sup>17,57,102</sup> Zhang *et al.* used a shape-memory epoxy resin as the bonding layer and low-surface-energy silica as the top layer to create a superamphiphobic coating with self-healing properties. The shape-memory bonding layer effectively drove the repair of damaged micro-/nanostructures in the superamphiphobic top layer (Fig. 5a). This coating exhibited excellent repetitive self-healing performance at the same location of scratches and was also capable of repairing more complex scratches, such as those in the shape of a star.<sup>103</sup>

Additionally, introducing elastic adhesives (e.g., polydimethylsiloxane and polyurethane) into superhydrophobic

coatings can not only enhance the adhesion but also impart elasticity to the coatings. This elasticity allows the coatings to deform under external impact or friction, reducing stress concentration and effectively improving the mechanical robustness of the coatings.<sup>40,89,104</sup> Furthermore, once the load is removed, the coating structure can recover to its original surface configuration. For example, Zhang *et al.* developed an elastic, mechanically robust superhydrophobic coating by combining polydimethylsiloxane with low-surface-energy silica nanoparticles (Fig. 5b). The tensile strength of this coating reached 4.7 MPa, demonstrating its excellent elasticity.<sup>89</sup> Thanks to its elasticity, the coating exhibited outstanding mechanical robustness, retaining excellent superhydrophobicity even after undergoing tape-peel tests, knife-scratching, and finger-wiping.

In summary, the mechanisms by which adhesives enhance the mechanical robustness of superhydrophobic coatings can be broadly categorized into three aspects: improving the adhesion between nanoparticles, as well as between the nanoparticles and the substrate; constructing microstructures that protect fragile nanostructures; and imparting functionality



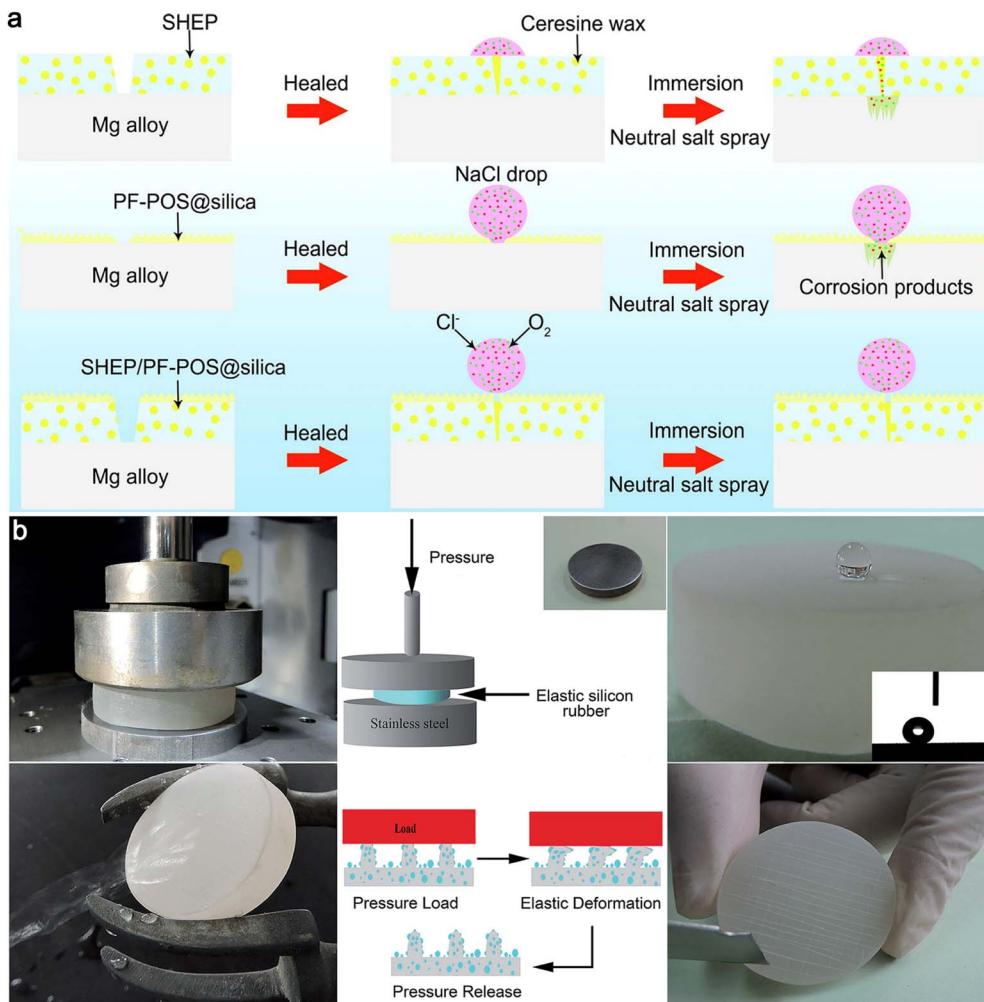


Fig. 5 (a) Schematic illustration of the mechanically robust superhydrophobic coating prepared using a self-healing adhesive. (b) Schematic illustration and photographs of the mechanically robust superhydrophobic coating prepared using an elastic adhesive. (a) Reproduced with permission from ref. 103. Copyright 2020, Elsevier. (b) Reproduced with permission from ref. 89. Copyright 2016, The Royal Society of Chemistry.

such as self-healing and elasticity. Each type of adhesive may contribute differently depending on its chemical nature and application method. A deeper understanding of these mechanisms aids in guiding the selection of suitable adhesives and the design of novel adhesives for the development of high-performance mechanically robust superhydrophobic coatings based on adhesives.

### 3. Progress of mechanically robust superhydrophobic coatings based on different types of adhesives

To date, extensive research has been conducted on the use of various types of adhesives in the fabrication of mechanically robust superhydrophobic coatings. This section provides a systematic overview of recent progress in the field. The frequently used adhesives are organic adhesives, inorganic adhesives, and solid powder adhesives (Table 1).

#### 3.1. Organic adhesives

Organic adhesives are widely employed in the fabrication of mechanically robust superhydrophobic coatings owing to their excellent film-forming capabilities and strong interfacial adhesion to a variety of substrates. Commonly used organic adhesives include epoxy resins, silicone resins, polyurethanes, silane-modified polyester resins, *etc.*

**3.1.1. Epoxy resins.** Epoxy resins are a class of thermosetting polymeric prepolymers or oligomers characterized by the presence of two or more reactive epoxy groups in their molecular structure.<sup>105</sup> Typically, these resins feature epoxy functionalities at both chain ends, with the polymer backbone incorporating aromatic rings, alkyl chains, hydroxyl groups, and ether linkages.<sup>105</sup> Based on their chemical composition, epoxy resins can be broadly categorized into glycidyl-type and non-glycidyl-type resins.<sup>105</sup> Epoxy resins have been extensively utilized in the fabrication of mechanically robust superhydrophobic coatings owing to their high mechanical strength and excellent adhesive properties across a wide range of



Table 1 Representative mechanically robust superhydrophobic coatings based on different adhesives<sup>a</sup>

Adhesive types	Nanoparticles	Mechanical robustness			Ref.	
		Testing methods	Cycles	CA and SA		
Organic adhesives	Epoxy resin	SiO <sub>2</sub> @CNTs	Tape peeling	300	CA >150°, SA = —	113
	Fluorinated epoxy resin	PTFE	Taber abrasion	100	CA >150°, SA <10°	114
	Silicone resin	SiO <sub>2</sub>	Sandpaper abrasion	50	CA = 155°, SA = 13°	118
	Fluorosilicone resin	SiO <sub>2</sub>	Sandpaper abrasion	30	CA >150°, SA = 16.1°	121
	Fluoride/fluorosilicone resin	TiO <sub>2</sub> @GO	Sandpaper abrasion	1000	CA >155°, SA <13°	119
	Polyurethane	SiO <sub>2</sub>	Tape peeling	100	CA >150°, SA <10°	130
	Fluorinated polyurethane	SiO <sub>2</sub>	Sandpaper abrasion	150	CA >150°, SA <10°	129
	Silicone-modified polyester	Cu-MOFs@attapulgite	Taber abrasion	150	CA >150°, SA <30°	87
	Acrylic resin	SiO <sub>2</sub>	Sandpaper abrasion	300	CA >155°, SA <20°	138
	SEBS	SiO <sub>2</sub>	Tape peeling	10	CA >150°, SA <5°	139
Inorganic adhesives	Aluminum phosphate adhesive	TiO <sub>2</sub>	Sandpaper abrasion	100	CA >150°, SA = —	146
	Portland cement adhesive	Diatomite, sand powder	Sandpaper abrasion	120	CA = 154°, SA = 8°	148
	Solid powder adhesives	Polystyrene microparticles	Chained SiO <sub>2</sub>	210	CA > 150°, SA <13°	59
	Polyester powder	PTFE	Sandpaper abrasion	100	CA >150°, SA <25°	151

<sup>a</sup> “—” means not mentioned.

substrates.<sup>72,106–112</sup> For instance, Zhang *et al.* synthesized hybrid nanoparticles by grafting silica nanoparticles onto the surface of carbon nanotubes, which were then incorporated into bisphenol A diglycidyl ether, and applied onto substrates *via*

a one-step spray-coating process (Fig. 6a).<sup>113</sup> The resulting superhydrophobic coating exhibited outstanding mechanical robustness, retaining its superhydrophobicity after 300 tape peeling cycles.

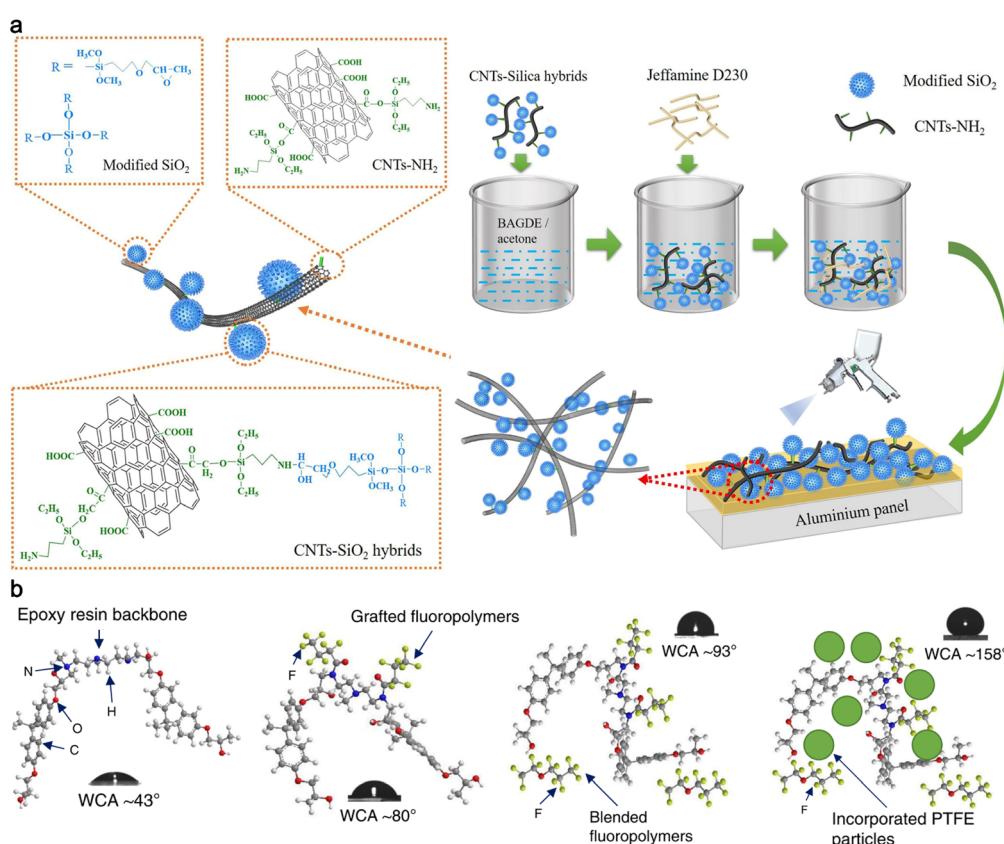


Fig. 6 Schematic illustration of the preparation of mechanically robust superhydrophobic coatings based on (a) an epoxy resin and (b) a fluorinated epoxy resin. (a) Reproduced with permission from ref. 113. Copyright 2021, Elsevier. (b) Reproduced with permission from ref. 114. Copyright 2018, Springer Nature.



Although the incorporation of epoxy resin can effectively enhance the mechanical robustness of superhydrophobic coatings, it plays a significant role in increasing the surface energy, resulting in insufficient superhydrophobicity, particularly under dynamic conditions. To address this issue, researchers have developed fluorinated epoxy resins by introducing fluorine groups to reduce the surface energy while retaining mechanical robustness.<sup>98,114</sup> For example, Peng *et al.* synthesized a fluorinated amine curing agent by reacting diethylenetriamine with heptafluorobutyric acid-functionalized fluoropolymers. This fluorinated amine was then used to graft low-surface-energy fluoropolymers onto the epoxy backbone, yielding a fluorinated epoxy resin. Finally, the fluorinated epoxy resin was blended with perfluoropolyether and polytetrafluoroethylene nanoparticles to fabricate a coating with excellent mechanical robustness and dynamic superhydrophobicity (Fig. 6b). The coating retained good

superhydrophobicity after 30 cycles of tape peeling or 100 cycles of Taber abrasion, and it also withstood high speed water jet impacts at velocities up to  $\sim 35 \text{ m s}^{-1}$ .<sup>114</sup>

In addition, shape memory epoxy resins have been utilized in the fabrication of superhydrophobic coatings with self-healing capabilities.<sup>102,103,115,116</sup> For example, Zhang *et al.* employed a shape memory epoxy resin as the bonding layer and fluorinated carbon nanotubes as the top layer, to develop a photothermal responsive self-healing superhydrophobic coating.<sup>116</sup> This coating demonstrated excellent self-healing performance, effectively restoring both the damaged micro-/nanostructures and surface chemistry.

**3.1.2 Silicone resins.** Silicone resins are a class of polymers characterized by a backbone formed by alternating silicon and oxygen atoms, with various organic groups bonded to the silicon atoms.<sup>117</sup> These resins contain both organic groups and inorganic structures, which contribute to their excellent

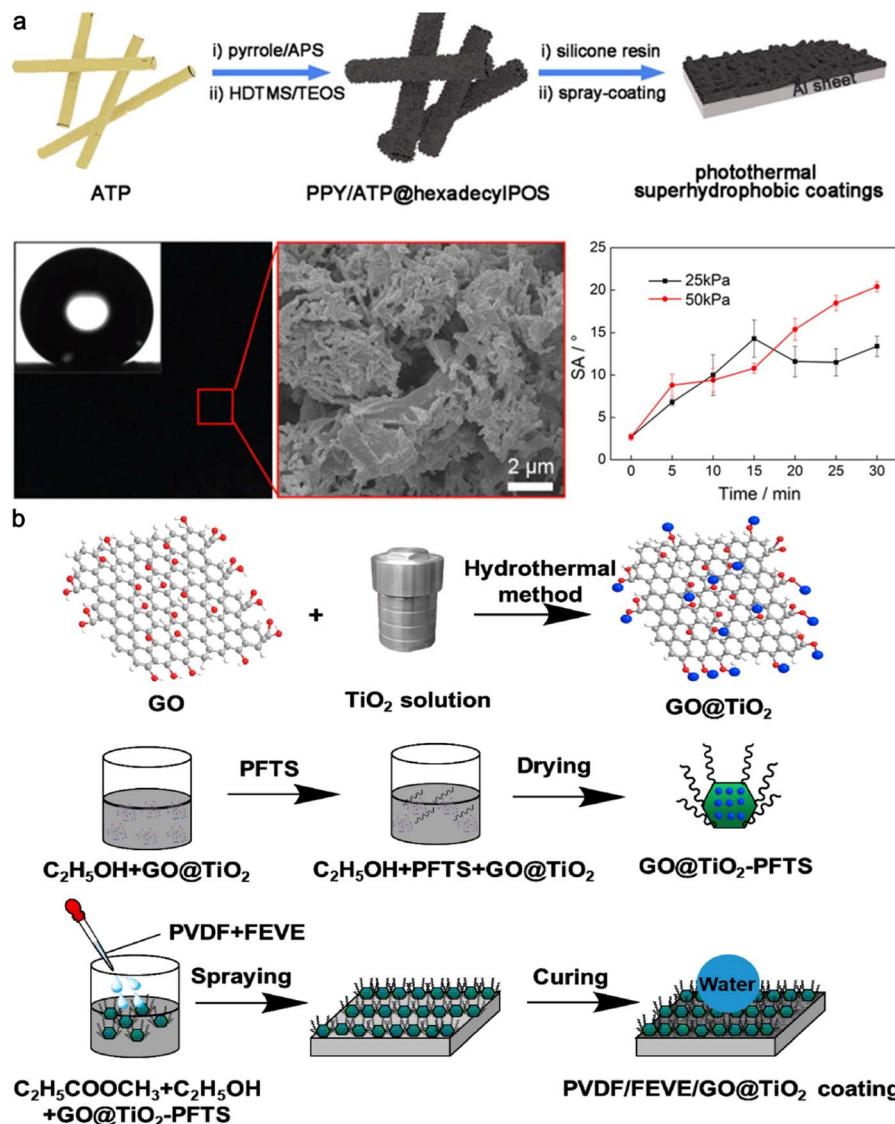


Fig. 7 (a) Schematic illustration, photographs, and mechanical robustness of the mechanically robust superhydrophobic coatings based on silicone resin. (b) Schematic illustration of the preparation of mechanically robust superhydrophobic coatings based on a fluorosilicone resin. (a) Reproduced with permission from ref. 69. Copyright 2022, Elsevier. (b) Reproduced with permission from ref. 119. Copyright 2021, Elsevier.



weather resistance and hydrophobic properties. Due to these advantages, silicone resins are widely used in the preparation of mechanically robust superhydrophobic coatings.<sup>68,118</sup> Chen *et al.* developed mechanically robust superhydrophobic coatings by combining silicone resins with low-surface-energy silica sol nanoparticles, using methods such as spraying and dip-coating.<sup>118</sup> The coating demonstrated remarkable superhydrophobicity after undergoing finger abrasion tests, knife scraping experiments, and 50 cycles of sandpaper abrasion. Furthermore, the coating exhibited extremely high hardness (pencil hardness of 9H) and excellent adhesion (5B). Xie *et al.* prepared a photothermal responsive, mechanically robust superhydrophobic coating on aluminum alloy surfaces by mixing silicone resins with low-surface-energy polypyrrole (PPY) modified attapulgite and applying a one-step spraying method. The coating exhibited a contact angle of up to 162.7° and a sliding angle as low as 2.7°. Even after exposure to water jet impacts at varying pressures, the coating retained excellent superhydrophobicity, demonstrating its exceptional mechanical robustness (Fig. 7a).<sup>69</sup>

In recent years, fluorosilicone resins have attracted attention due to their low surface energy and chemical inertness.<sup>120</sup> Fluorosilicone resins are synthetic resins formed by blending fluororesins with silicone resins. Compared to silicone resins, fluorosilicone resins possess a lower surface energy, and the contact angle of water on the film typically exceeds 90°. They have also been used to fabricate mechanically robust superhydrophobic coatings.<sup>119–123</sup> Liu *et al.* grew TiO<sub>2</sub> nanoparticles *in situ* on the surface of graphene oxide (GO) *via* a hydrothermal reaction, followed by low surface energy modification. The resulting composite was then incorporated into a polyvinylidene fluoride/fluorosilicone resin adhesive to fabricate a mechanically robust superhydrophobic coating (Fig. 7b). This coating retained its superhydrophobicity after 1000 cycles of sandpaper abrasion, demonstrating outstanding mechanical robustness.<sup>119</sup>

**3.1.3 Polyurethanes.** Polyurethane adhesives refer to adhesives that contain urethane groups in their molecular chains, which are formed through the reaction of isocyanates and polyols during the polymerization process.<sup>124</sup> Due to the presence of these urethane groups, polyurethane adhesives exhibit high reactivity and polarity, resulting in excellent adhesion to a wide range of substrates.<sup>124–127</sup> Additionally, polyurethane imparts flexibility to superhydrophobic coatings, further enhancing their mechanical robustness.<sup>128–131</sup> Zhi *et al.* mixed polyurethane with low-surface-energy silica nanoparticles, and by a one-step spraying method, fabricated a mechanically robust superhydrophobic coating.<sup>130</sup> This coating demonstrated outstanding mechanical robustness, retaining a sliding angle of less than 10° after 100 cycles of tape peeling.

To further minimize the impact of polyurethane adhesives on the superhydrophobicity of the coatings, low-surface-energy fluorinated polyurethane adhesives were synthesized and used to fabricate mechanically robust superhydrophobic coatings.<sup>132,133</sup> For example, Fu *et al.* synthesized fluorinated polyurethane through a two-step “click” chemistry reaction,

incorporated low-surface-energy silica nanoparticles, and applied the composite to a substrate *via* spraying (Fig. 8a).<sup>129</sup> The resulting coating retained its excellent superhydrophobicity after 150 cycles of sandpaper abrasion.

**3.1.4 Silicone-modified polyester resins.** Silicone-modified polyester resins, synthesized by reacting silicone with polyester, offer good hydrophobicity, weather resistance, and mechanical strength. We have developed a series of coatings based on this type of adhesive that exhibit both excellent mechanical robustness and dynamic superhydrophobicity.<sup>86–88,134–136</sup> For instance, by combining silicone-modified polyester adhesive with low-surface-energy Cu-MOFs@attapulgite nanoparticles and using ethanol as a non-solvent to induce phase separation of the adhesive, we successfully fabricated a mechanically robust superhydrophobic coating (Fig. 8b).<sup>87</sup> Phase separation degree and adhesive content significantly influence the coatings' properties by regulating the structural parameters and morphology. In simulated/real icing environments, the coatings simultaneously show (i) high superhydrophobicity and stable Cassie–Baxter states due to their low-surface energy, three-tier micro-/nano-/nanostructure, (ii) excellent photothermal effect primarily due to nanosized MOFs, and (iii) good mechanical robustness by the phase-separated adhesive, reinforcement with attapulgite and the coatings' self-similar structure. This coating retained its outstanding superhydrophobicity after 150 cycles of Taber abrasion or 300 cycles of tape peeling.

**3.1.5 Other organic adhesives.** In addition to the adhesives mentioned above, poly(methyl methacrylate), acrylic resins, polyurea, block copolymers, *etc.*, have also been used for the fabrication of mechanically robust superhydrophobic coatings.<sup>137–143</sup> For example, Li *et al.* developed a superhydrophobic coating with outstanding mechanical robustness by combining water-based acrylic copolymers with low-surface-energy silica nanoparticles.<sup>138</sup> The coating retained excellent superhydrophobicity after 300 cycles of sandpaper abrasion. Similarly, Wang *et al.* employed a simple one-step spraying method to fabricate a mechanically robust superhydrophobic coating based on polystyrene-*b*-poly(ethylene-*co*-butylene)-*b*-polystyrene (SEBS) adhesive.<sup>139</sup> This coating demonstrated remarkable superhydrophobicity after undergoing tape peeling and sand impact tests.

## 3.2 Inorganic adhesives

Compared to organic adhesives, inorganic adhesives offer distinct advantages, including low toxicity, high adhesion strength, excellent solvent resistance, high thermal stability, and superior aging resistance.<sup>144</sup> Moreover, many inorganic adhesives are water-soluble, minimizing the reliance on organic solvents and enhancing environmental compatibility.<sup>144</sup> Among these, phosphate- and silicate-based inorganic adhesives have been successfully employed in the fabrication of mechanically robust superhydrophobic coatings.<sup>144–149</sup>

**3.2.1 Phosphate adhesives.** Phosphate adhesives contain numerous hydroxyl groups and unsaturated P=O bonds, which enable the formation of crosslinked structures through



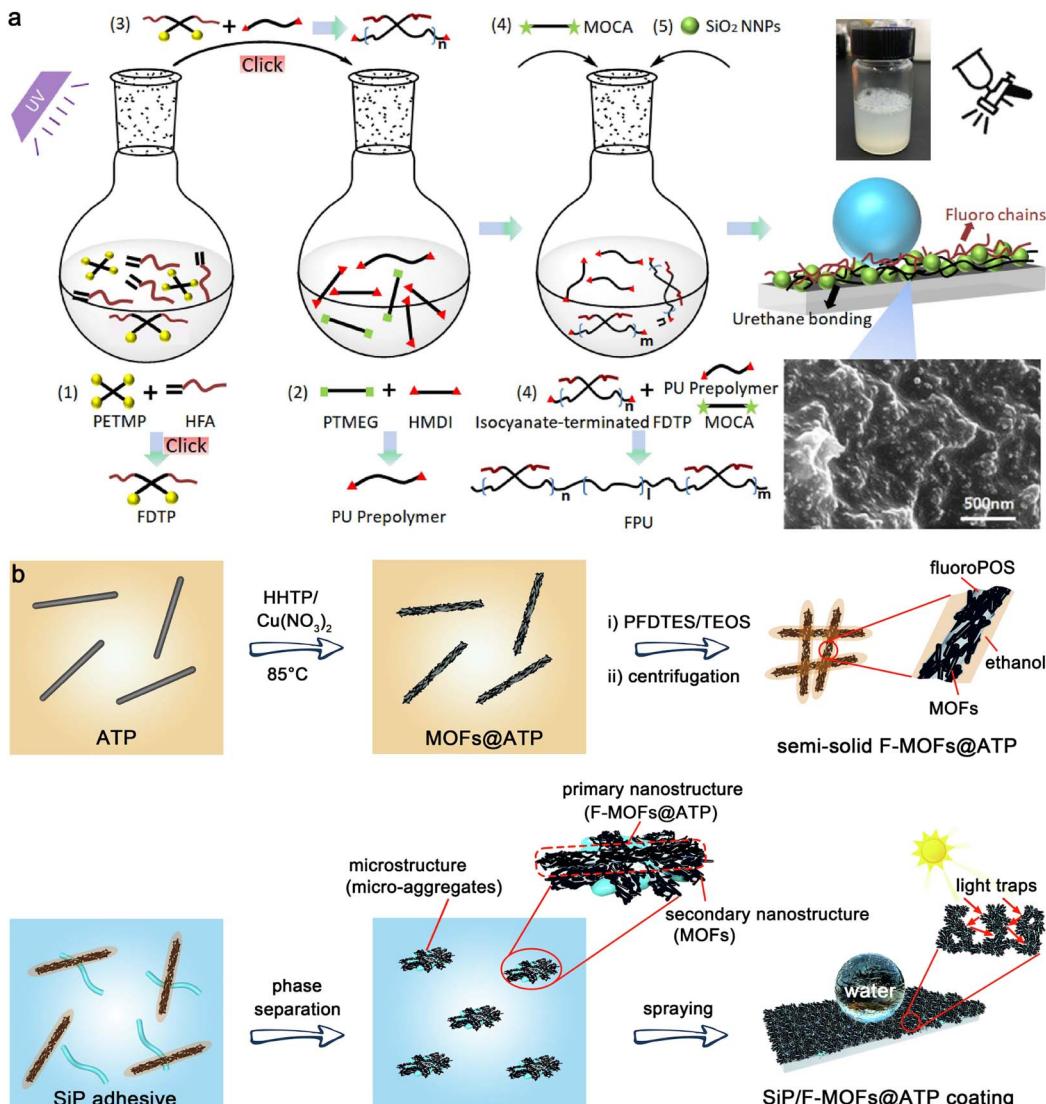


Fig. 8 Schematic illustration of the preparation mechanically robust superhydrophobic coatings based on (a) fluorinated polyurethane and (b) silicone-modified polyester adhesives. (a) Reproduced with permission from ref. 129. Copyright 2021, Elsevier. (b) Reproduced with permission from ref. 87. Copyright 2024, Springer Nature.

dehydration during condensation reactions. The hydroxyl groups in phosphate adhesives can form hydrogen bonds or metal–oxygen ion complexes with ceramic or metal substrates, thereby enhancing adhesion.<sup>147,149</sup> Liu *et al.* incorporated low-surface-energy  $\text{TiO}_2$  nanoparticles into an aluminum phosphate adhesive and sprayed the mixture onto a substrate, followed by high temperature curing.<sup>146</sup> The resulting superhydrophobic coating exhibited outstanding superhydrophobicity after 100 cycles of sandpaper abrasion or 50 cycles of sand impact (Fig. 9a and b).

**3.2.2 Silicate inorganic adhesives.** In addition to phosphate adhesives, silicate adhesives are another type of inorganic adhesive used to prepare mechanically robust superhydrophobic coatings. Silicate inorganic adhesives offer the advantage of curing at room temperature, which addresses the high temperature curing requirement of phosphate adhesives.

Wang *et al.* developed a mechanically robust superhydrophobic surface by combining a Portland cement-based adhesive with diatomaceous earth and sand powder. This coating retained its excellent superhydrophobicity after 120 cycles of sandpaper abrasion (Fig. 9c).<sup>148</sup>

### 3.3 Solid powder adhesives

While the introduction of liquid adhesives can effectively improve the mechanical robustness of superhydrophobic coatings, it also leads to the embedding of low-surface-energy nanoparticles, which causes a sharp increase in the surface energy and reduction in the roughness of superhydrophobic coatings.<sup>86</sup> The use of solid powder adhesives to prepare mechanically robust superhydrophobic coatings can effectively address this issue.<sup>150</sup> Yu *et al.* mixed polystyrene microparticles with low-surface-energy chained nanoparticles and sprayed the



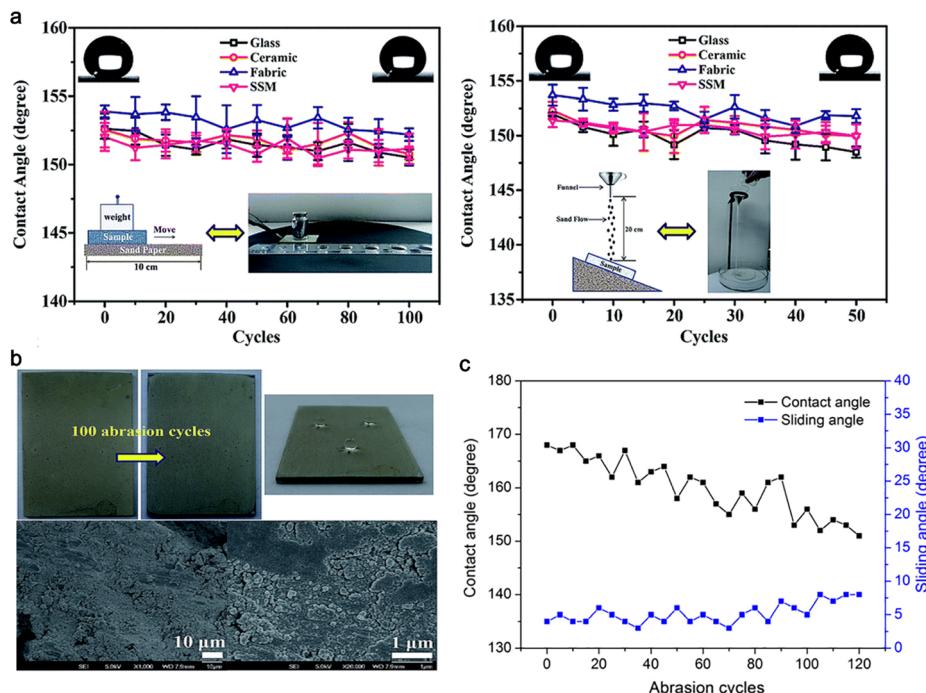


Fig. 9 (a) Mechanical robustness of the superhydrophobic coatings based on phosphate inorganic adhesives. (b) Photographs and SEM images of the superhydrophobic coatings based on phosphate inorganic adhesives after abrasion. (c) Mechanical robustness of the superhydrophobic coatings based on silicate inorganic adhesives. (a and b) Reproduced with permission from ref. 146. Copyright 2017, The Royal Society of Chemistry. (c) Reproduced with permission from ref. 148. Copyright 2019, Elsevier.

mixture onto a substrate surface. By combining a hot-pressing process at 180 °C and 1 kPa pressure, they developed a mechanically robust superhydrophobic coating (Fig. 10a).<sup>59</sup> This coating demonstrated excellent superhydrophobicity and mechanical robustness, with a contact angle greater than 150° and a sliding angle that only increased to 13.2° after 1500 cm of abrasion under a pressure of 10 kPa.

In addition, solid powder adhesives have also been used to prepare superhydrophobic powder coatings.<sup>152–154</sup> These coatings are produced by mixing powder adhesives with low-surface-energy nanoparticles, electrostatically spraying the mixture onto a substrate, and curing at high temperatures. Commonly used powder adhesives in such systems include polyester and polysulfone. Huang *et al.* mixed polyester powder adhesive with polytetrafluoroethylene nanoparticles, electrostatically sprayed the mixture onto a substrate, and cured it at 200 °C for 15 min to create a mechanically robust superhydrophobic coating (Fig. 10b).<sup>151</sup> The coating retained excellent superhydrophobicity after 100 cycles of sandpaper abrasion or tape peeling.

In conclusion, different types of adhesives can effectively enhance the mechanical robustness of superhydrophobic coatings. According to statistics from the Web of Science, organic adhesives are widely used by researchers to prepare mechanically robust superhydrophobic coatings, accounting for as much as 84.3% (Fig. 11a). Among these organic adhesives, epoxy resins, polyurethanes, and silicone resins are the most used, accounting for 34.7%, 34.0%, and 20.8%, respectively (Fig. 11b). While significant progress has been made in the

development of mechanically robust superhydrophobic coatings based on different types of adhesives, each type of adhesive still has its limitations in practical use. For example, organic adhesives often suffer from inadequate long-term weather resistance and environmental sustainability; inorganic adhesives have high surface energy, resulting in poor superhydrophobicity; solid powder adhesives require high temperature curing, and so on. Therefore, future efforts should focus on the design and synthesis of ideal adhesives that combine excellent long-term weather resistance, low surface energy, high mechanical strength, strong adhesion, water-based formulations, room-temperature curing, and other desirable properties, or on the development of hybrid adhesive systems to address these shortcomings. This will effectively promote the development and practical application of mechanically robust superhydrophobic coatings based on adhesives.

In addition to adhesives, low-surface-energy nanoparticles are another key component in the preparation of mechanically robust superhydrophobic coatings based on adhesives. The morphology and size of these nanoparticles significantly influence both superhydrophobicity and mechanical robustness of the coatings.<sup>83,84</sup> To date, the nanoparticles most commonly employed in the preparation of such coatings include inorganic oxide nanoparticles (e.g., SiO<sub>2</sub>, TiO<sub>2</sub>, ZnO and Al<sub>2</sub>O<sub>3</sub>),<sup>143,155–162</sup> carbon-based nanoparticles (e.g., carbon nanotubes, carbon black and GO),<sup>102,119,163–166</sup> and clay mineral nanoparticles (e.g., attapulgite, halloysite, montmorillonite and diatomite).<sup>83,87,88,167</sup> Among these, inorganic oxide nanoparticles are particularly favoured due to their high hardness and excellent

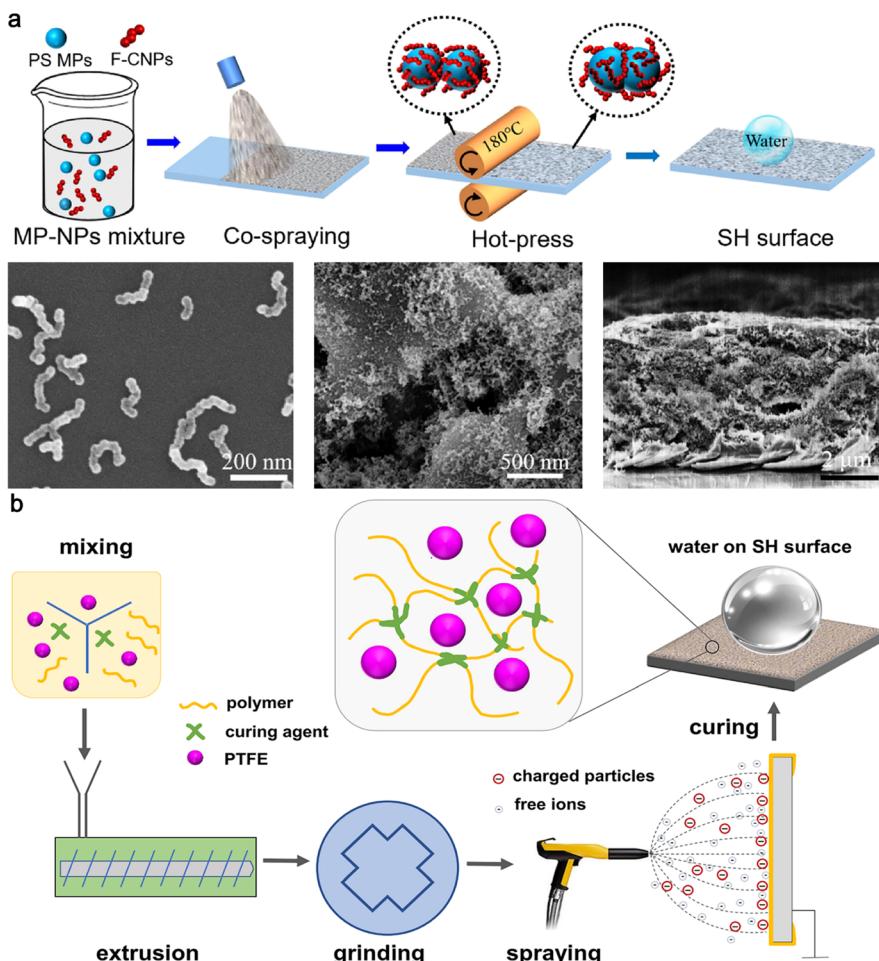


Fig. 10 (a) Schematic illustration and SEM images of the mechanically robust superhydrophobic coatings based on solid powder adhesive. (b) Schematic illustration of the mechanically robust superhydrophobic powder coatings. (a) Reproduced with permission from ref. 59. Copyright 2021, Elsevier. (b) Reproduced with permission from ref. 151. Copyright 2021, American Chemical Society.

chemical stability.<sup>75</sup> Of the inorganic oxides,  $\text{SiO}_2$  nanoparticles are especially popular due to their large specific surface area and surface-rich hydroxyl groups (Si-OH), which facilitate functionalization with low surface energy materials such as alkyl silanes.<sup>51,74</sup>

The size of nanoparticles also plays a crucial role in determining superhydrophobicity and mechanical robustness of the coatings.<sup>48,51,83,84,168</sup> Smaller nanoparticles can form richer micro-/nanostructures, which significantly enhance

superhydrophobicity of the coatings.<sup>51</sup> However, as the size of nanoparticles decreases, challenges related to their uniform dispersion within the adhesive matrix become more pronounced.<sup>51</sup> While larger nanoparticles are easier to disperse, they fail to form the rich micro-/nanostructures necessary for achieving superior superhydrophobicity.<sup>51</sup> Furthermore, the size of the nanoparticles also affects the mechanical robustness of the coatings. While smaller nanoparticles can contribute to richer micro/nanostructures, these structures are more

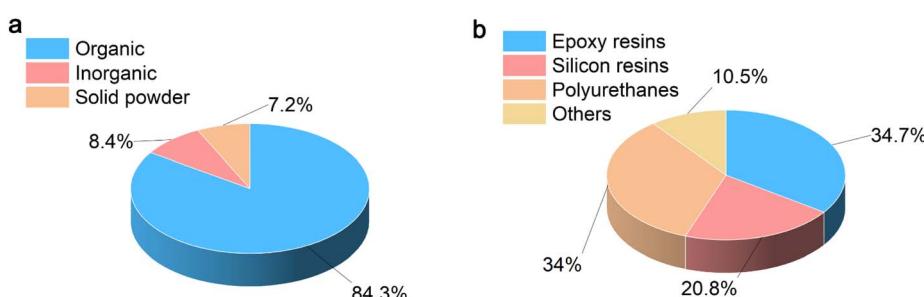


Fig. 11 Proportion of articles of mechanically robust superhydrophobic coatings based on (a) different types of adhesives and (b) different types of organic adhesives as of December 2024.

susceptible to damage under external stress, resulting in poor mechanical robustness of the coatings.<sup>169</sup> Therefore, selecting nanoparticles of the appropriate size is crucial for preparing mechanically robust superhydrophobic coatings with optimal comprehensive performance.

Despite significant progress in using various low-surface-energy nanoparticles to prepare mechanically robust superhydrophobic coatings based on adhesives, several challenges remain. One primary issue is ensuring the uniform dispersion of these nanoparticles within the adhesive matrix, as this directly influences both the superhydrophobicity and mechanical robustness of the coatings. Additionally, the long-term performance stability of the nanoparticles within the adhesive matrix is another critical challenge. Therefore, future research should focus on solving these two issues.

## 4. Progress of mechanically robust superhydrophobic coatings *via* different adhesive application methods

In addition to the chemical properties and molecular structures of adhesives, their application methods within the coating system play a critical role in determining the mechanical robustness of superhydrophobic coatings. Currently, these application methods can be broadly categorized into three types: direct blending of adhesives with nanoparticles, blending of adhesive microspheres formed by phase separation with nanoparticles, and the application of adhesives as an interfacial bonding layer (Table 2).

### 4.1 Direct blending of adhesives with nanoparticles

Direct blending of adhesives with nanoparticles refers to the method where adhesives are directly mixed with low-surface-energy nanoparticles and then applied to a substrate surface

through techniques such as spraying, brushing or dipping.<sup>72,86,102,160,161,163,170–172</sup> This approach is simple to implement, making it suitable for large-scale production, and it is the most frequently used method for adhesives in superhydrophobic coatings.<sup>53</sup> For example, Xue *et al.* demonstrated a direct mixing of acrylic resin with low-surface-energy silica nanoparticles, which were then sprayed onto a substrate to form a mechanically robust superhydrophobic coating.<sup>131</sup> The resulting coating retained excellent superhydrophobicity after 200 cycles of sandpaper abrasion. Similarly, Gu *et al.* developed mechanically robust superhydrophobic coatings by mixing porous diatomite loaded with low-surface-energy silica nanoparticles and adhesives (e.g., epoxy resin, acrylic resin and polyurethane), followed by spraying (Fig. 12a).<sup>65</sup> These coatings preserved their superior superhydrophobicity even after 1000 cycles of Taber abrasion.

It is important to note that the ratio of low-surface-energy nanoparticles to adhesives in this method plays a crucial role in determining the performance of superhydrophobic coatings, as excessive adhesives can significantly increase the surface energy of the coatings, thereby compromising the superhydrophobicity.<sup>155,166</sup> Zhang *et al.* systematically investigated the effect of the ratio of low-surface-energy nanoparticles to adhesives on both the superhydrophobicity and mechanical robustness of the coatings (Fig. 12b). The results revealed that as the ratio of low-surface-energy nanoparticles to adhesives increased, the superhydrophobicity of the coating rapidly improved before stabilizing, while the mechanical robustness initially increased and then decreased.<sup>173</sup>

### 4.2 Blending of adhesive microspheres with nanoparticles

To address the limitations (e.g., poor superhydrophobicity) of the direct blending of adhesives with nanoparticles in preparing mechanically robust superhydrophobic coatings, researchers

Table 2 Representative mechanically robust superhydrophobic coatings based on different adhesive application methods<sup>a</sup>

Adhesive types	Nanoparticles	Adhesive application methods	Mechanical robustness				Ref.
			Testing methods	Cycles	CA and SA		
Acrylic resin	SiO <sub>2</sub>	Direct blending of adhesives with nanoparticles	Sandpaper abrasion	200	CA >150°, SA <60°		131
Epoxy resin <i>etc.</i>	Diatomite@SiO <sub>2</sub>		Taber abrasion	1000	CA >150°, SA <30°		65
Silicone-modified polyester	SiO <sub>2</sub> , carbon black		Taber abrasion	300	CA >150°, SA <40°		86
Silicone-acrylic emulsion	Halloysite@SiO <sub>2</sub>		Sandpaper abrasion	50	CA = 157°, SA = 6°		176
Ethylene tetrafluoroethylene	Polyaniline/CNTs		Taber abrasion	8000	CA = 149°, SA = —		141
Silicone-modified polyester	SiO <sub>2</sub>	Blending of adhesive microspheres with nanoparticles	Paper abrasion	100	CA > 150°, SA < 15°		136
Polyolefin	SiO <sub>2</sub>		Sandpaper abrasion	300	CA = 159.9°, SA = 7.5°		64
Poly(styrene- <i>co</i> -isoprene)	SiO <sub>2</sub>		Sandpaper abrasion	30	CA = 165°, SA = 3°		175
Silicone-modified polyester	Attapulgite		Taber abrasion	120	CA >150°, SA <30°		88
Polyolefin	SiO <sub>2</sub>		Taber abrasion	200	CA >150°, SA = 5°		177
Polyurethane	SiO <sub>2</sub>	Interfacial bonding layer	Sandpaper abrasion	150	CA = 150.1°, SA = 35.8°		74
Polyurethane	SiO <sub>2</sub>		Taber abrasion	400	CA = 154.8°, SA = 4.3°		178
Epoxy resin	Fe <sub>3</sub> O <sub>4</sub> @SiO <sub>2</sub>		Sandpaper abrasion	100	CA > 160°, SA = —		179
Double-sided tapes	TiO <sub>2</sub>		Sandpaper abrasion	40	CA >156°, SA = —		66
Epoxy resin	Cellulose, SiO <sub>2</sub>	Direct blending + interfacial bonding layer	Taber abrasion	3000	CA >150°, SA <10°		173

<sup>a</sup> “—” means not mentioned.



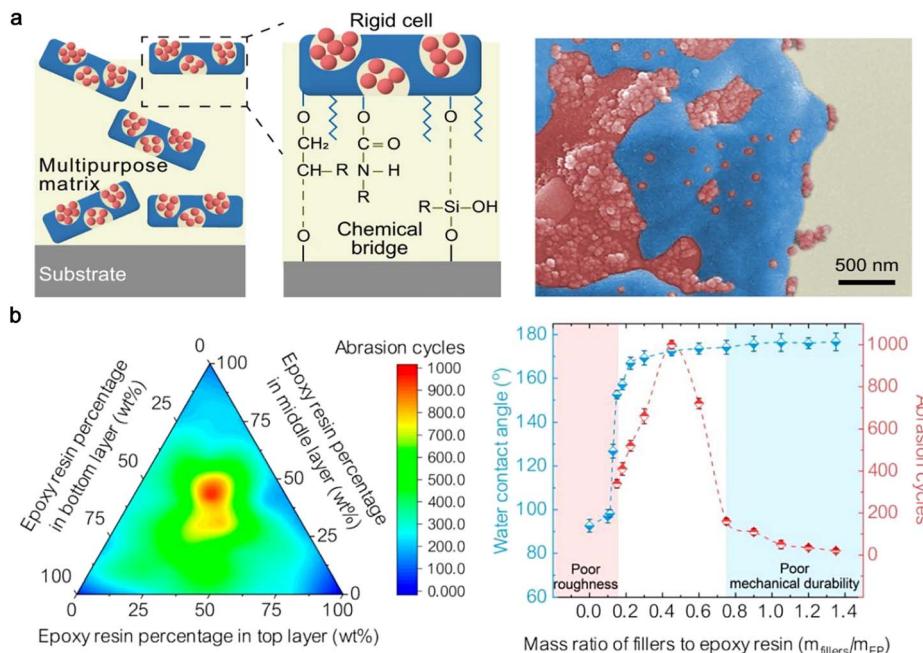


Fig. 12 (a) Schematic illustration and SEM image of the cellular superhydrophobic coatings prepared by mixing porous diatomite loaded with low-surface-energy silica nanoparticles with adhesives. (b) Effects of mass ratio of nanoparticles to adhesives on superhydrophobicity and mechanical robustness of the superhydrophobic coatings prepared by the direct blending of adhesives with nanoparticles. (a) Reproduced with permission from ref. 65. Copyright 2023, Springer Nature. (b) Reproduced with permission from ref. 173. Copyright 2022, WILEY-VCH.

have developed the blending of adhesive microspheres with nanoparticles. In this method, the adhesive is first induced to phase separation in a nonsolvent, forming adhesive microspheres, which are then mixed with low-surface-energy nanoparticles.<sup>87,88,134,174</sup> This strategy allows the low-surface-energy nanoparticles to adhere to the surface of the adhesive microspheres, forming a composite of adhesive/low-surface-energy nanoparticle microspheres. This effectively resolves the issue of significantly increased surface energy caused by the direct blending of adhesives with nanoparticles, thereby enhancing the mechanical robustness of the coating while minimizing the impact on its superhydrophobicity.<sup>135,136,175</sup> Wei *et al.* used ethanol as the nonsolvent to induce phase separation in a polyolefin adhesive, which was then combined with low-surface-energy silica nanoparticles to create a superhydrophobic coating with both excellent dynamic superhydrophobicity and mechanical robustness (Fig. 13a).<sup>64</sup> After 20 h of simulated rainfall, the coating retained its original superhydrophobicity, and after 300 cycles of sandpaper abrasion, its contact angle remained greater than 150° with a sliding angle less than 10°. Thanks to its simple preparation process and outstanding overall performance, this coating has also been successfully applied for prevention of rain attenuation of 5G/weather radomes.

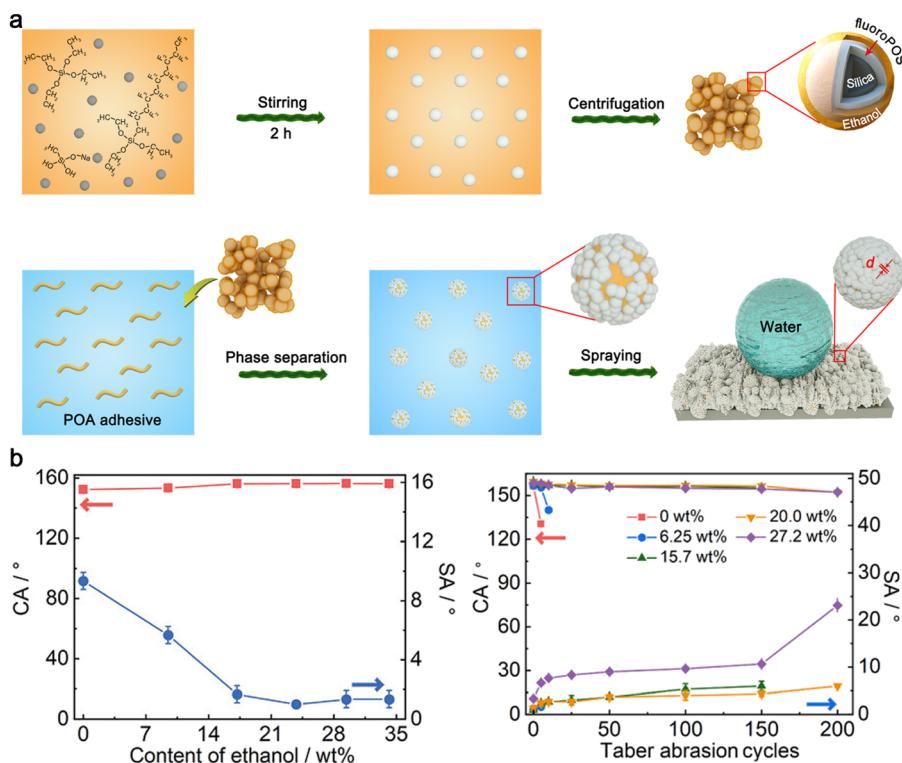
It is important to note that the degree of phase separation and the content of adhesive are key factors influencing the performance of superhydrophobic coatings in this application method.<sup>180,181</sup> Li *et al.* systematically investigated the effects of phase separation degree and the content of adhesive on both superhydrophobicity and mechanical robustness of the

coatings (Fig. 13b).<sup>177</sup> The results revealed that as the phase separation degree increased, the superhydrophobicity initially improved before stabilizing, while the mechanical robustness increased before eventually decreasing. Additionally, superhydrophobicity decreased with increasing adhesive content, while mechanical robustness initially increased before declining.

#### 4.3 Interfacial bonding layer

An alternative effective strategy to enhance the mechanical robustness of superhydrophobic coatings is the use of a layered design approach, where the adhesive is used as a separate interfacial bonding layer. This strategy significantly improves the adhesion between the coating and the substrate without affecting the surface energy of the superhydrophobicity coating.<sup>178,182-189</sup> Li *et al.* developed a mechanically robust superhydrophobic coating by combining a water-based polyurethane interfacial bonding layer with a silica superhydrophobic coating.<sup>74</sup> Thanks to the bonding layer, the coating retained its excellent superhydrophobicity after 150 cycles of sandpaper abrasion or 200 cycles of tape peeling (Fig. 14a). In addition to using adhesives as interfacial bonding layers, commercially available double-sided tapes have also been employed as interfacial bonding layers to create mechanically robust superhydrophobic coatings.<sup>190</sup> Lu *et al.* demonstrated that using double-sided tape as the interfacial bonding layer significantly improved the mechanical robustness of superhydrophobic coatings.<sup>66</sup> After being rubbed with a finger, scratched with a knife, or abrasion with sandpaper for 40 cycles, the coating retained its exceptional superhydrophobicity.





**Fig. 13** (a) Schematic illustration of the superhydrophobic coatings prepared by blending adhesive microspheres with nanoparticles. (b) Effects of phase separation degree on superhydrophobicity and mechanical robustness of the superhydrophobic coatings prepared by blending adhesive microspheres with nanoparticles. (a) Reproduced with permission from ref. 64 Copyright 2023, Springer Nature. (b) Reproduced with permission from ref. 177. Copyright 2024, WILEY-VCH.

While the introduction of an interfacial bonding layer effectively enhances the adhesion between the coating and the substrate, it has minimal impact on improving the internal cohesion of the superhydrophobic coating, resulting in only limited enhancement of mechanical robustness. To address this issue, researchers have combined the direct blending method with the interfacial bonding layer approach to develop mechanically robust superhydrophobic coatings.<sup>173,191</sup> For example, Zhang *et al.*, inspired by the structure of human skin, designed a superhydrophobic coating with a three-layer structure that exhibited excellent mechanical robustness (Fig. 14b).<sup>173</sup> The authors first sprayed epoxy resin onto a substrate to form the interfacial bonding layer; then, epoxy resin was used as the main component, with a small amount of low-surface-energy nanoparticles introduced as a transition layer; finally, a mixture of a small amount of epoxy resin and low-surface-energy nanoparticles was applied as the top layer to create a mechanically robust superhydrophobic coating. The coating retained excellent superhydrophobicity after 3000 cycles of Taber abrasion, with a contact angle greater than 150° and a sliding angle less than 10°.

In summary, although the direct blending of adhesives with nanoparticles can effectively enhance the mechanical robustness of superhydrophobic coatings, it often results in high surface energy, thereby compromising dynamic superhydrophobicity. The blending of adhesive microspheres with nanoparticles, in which phase separation is induced by

nonsolvents to expose low-surface-energy components, can effectively mitigate this issue. However, the resulting mechanical enhancement is limited due to the encapsulation of the adhesive by low-surface-energy nanoparticles. Similarly, using adhesives as an interfacial bonding layer provides moderate improvement in mechanical robustness but remains insufficient. To address these limitations, recent studies have explored hybrid strategies that combine direct blending with bonding layer, achieving notable improvements in mechanical robustness. Nonetheless, these coatings still exhibit suboptimal dynamic superhydrophobicity. Consequently, future research should aim to further optimize the integration of various adhesive application methods to construct advanced superhydrophobic coatings that concurrently deliver exceptional mechanical robustness and dynamic superhydrophobicity.

## 5. Commercial status of mechanically robust superhydrophobic coatings based on adhesives

Although significant progress has been made in laboratory research on mechanically robust superhydrophobic coatings based on adhesives, their true entry into industrial applications and the market still faces high barriers. Compared to basic research, commercialized superhydrophobic coatings must strike a balance across several dimensions, including



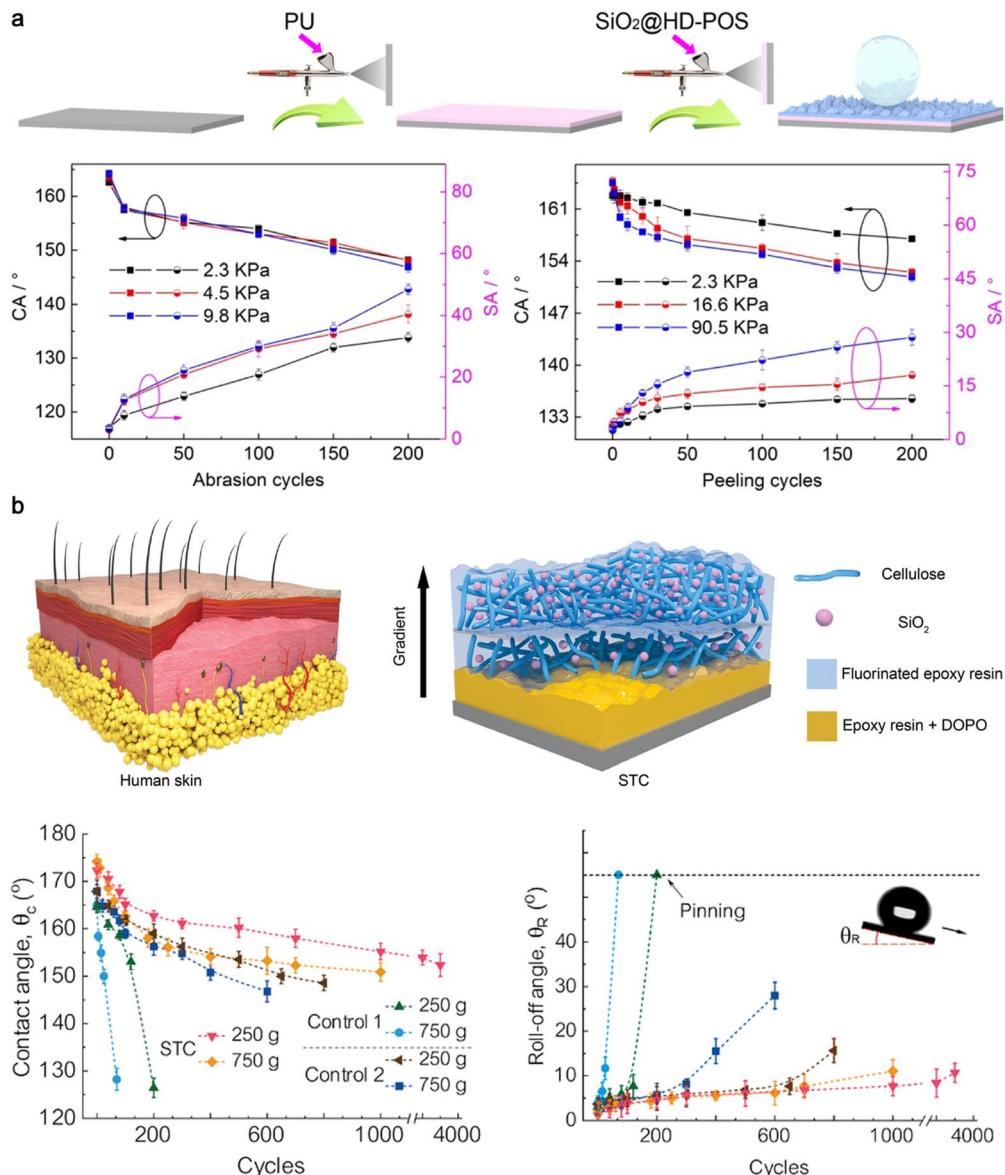


Fig. 14 Schematic illustration and mechanical robustness of the superhydrophobic coatings based on (a) the adhesive bonding layer and (b) a hybrid strategy combining direct blending and adhesive bonding layer. (a) Reproduced with permission from ref. 74. Copyright 2018, American Chemical Society. (b) Reproduced with permission from ref. 173. Copyright 2022, WILEY-VCH.

performance stability, ease of application, environmental compatibility, and cost control.

According to our statistics, there are currently 13 commercially available mechanically robust superhydrophobic coating products based on adhesives from 9 different companies, as detailed in Table 3. These products primarily originate from China and the United States. Among them, mechanically robust superhydrophobic coating products based on adhesives that use adhesives as coating-substrate interfacial bonding layers dominate the market, accounting for 76.9% of the products. Notable examples include NeverWet<sup>®</sup> and Ultra-Ever Dry<sup>®</sup>. There are two mechanically robust superhydrophobic coatings where adhesives are applied by direct blending: NANOMYTE SuperCN Plus by NEI Corporation and XN-FSSL by Shandong

Xinna Superhydrophobic New Materials LLC. Additionally, there is one mechanically robust superhydrophobic coating where adhesives are applied by formed adhesive microspheres: XN-FSSA, also produced by Shandong Xinna Superhydrophobic New Materials LLC.

In terms of mechanical robustness, the most outstanding product is XN-FSSL by Shandong Xinna Superhydrophobic New Materials LLC, which retains its superhydrophobicity even after 300 cycles of sandpaper abrasion. In addition to mechanical robustness, the company's superhydrophobic coating products also exhibit excellent dynamic superhydrophobicity and weather resistance, making them the current market leaders in terms of overall performance for mechanically robust superhydrophobic coatings based on adhesives.



Table 3 Commercialized mechanically robust superhydrophobic coating products based on adhesives<sup>a</sup>

Products	Country	Adhesive application method	Mechanical robustness	Company and website
NeverWet®	USA	Interfacial bonding layer	—	192
Ultra-Ever Dry®	USA		After 30 cycles of Taber abrasion, superhydrophobic	193
CytoThane TS	USA		After 10 cycles of Taber abrasion, superhydrophobic	194
JN-SS002/JN-SS003	China		—	195
DSAN-S2001-I	China		—	196
DSAN-S2001-II			—	
DSAN-S2001-III			—	
HIREC® 100	Japan		—	197
HIREC® 300 W			—	
NANOMYTE® SuperCN Plus	USA	Direct blending of adhesives with nanoparticles	—	198
SPN-64	China		—	199
XN-FSSL	China		After 300 cycles of sandpaper abrasion, CA >150°, SA = 7.5°	200
XN-FSSA	China	Blending of adhesive microspheres with nanoparticles	After 50 cycles of Taber abrasion, CA >150°, SA <10°	200

<sup>a</sup> “—” means not mentioned.

## 6. Common testing methods for mechanical robustness of superhydrophobic coatings

Superhydrophobic coatings are often subjected to external environmental factors during practical applications, leading to damage of their surface structure, which in turn causes the coatings to lose superhydrophobicity.<sup>64</sup> Therefore, assessing the mechanical robustness of superhydrophobic coatings has become a critical factor for their widespread application.<sup>46</sup> In recent years, several testing methods have been developed and widely applied to evaluate the mechanical robustness of superhydrophobic coatings. These methods mainly include abrasion, tape peeling, and sand impact.<sup>46,53,87</sup>

### 6.1 Abrasion tests

The surface structure of superhydrophobic coatings may be damaged by touch or abrasion. To simulate this damage scenario, abrasion test is employed to evaluate the mechanical robustness of the coatings. The most used abrasion testing methods include linear reciprocating abrasion and Taber abrasion tests.

The linear reciprocating abrasion test is one of the most used methods for evaluating the mechanical robustness of superhydrophobic coatings. In this test, a coating sample is placed in contact with an abrasive material (e.g., sandpaper, rubber, etc.) under a vertical load, and a reciprocating motion along a straight path is applied to the coating surface.<sup>74,201</sup> One complete reciprocating stroke is defined as one cycle. By monitoring the changes in superhydrophobicity and surface morphology of the coating after a specific number of cycles, the mechanical robustness of the coating can be quantitatively evaluated. The core evaluation metric is the total number of

abrasion cycles or the total abrasion distance required for the coating to lose its superhydrophobicity.

The Taber abrasion test utilizes a Taber type abrasion tester for evaluation. This device consists of a rotating platform and a pair of standard abrasive wheels (e.g., CS-0, CS-10, etc.). The coating sample is fixed onto the rotating platform, and the abrasive wheels come into contact with the coating under a specific load, generating abrasive action as the platform rotates.<sup>86,87</sup> One complete rotation of the platform is defined as one cycle. By analyzing the changes in the coating's superhydrophobicity, mass loss, thickness reduction, and surface morphology during the abrasion process, the mechanical robustness of the coating is quantitatively evaluated. The core evaluation metric is the maximum number of cycles that the coating can withstand while retaining its superhydrophobicity. Compared to the linear reciprocating abrasion test, this method shows better reproducibility and comparability.

### 6.2 Tape peeling test

The tape peeling test is primarily used to simulate the peeling and detachment of coatings during service due to external environmental factors. In this method, a standard pressure-sensitive adhesive tape (e.g., 3M Scotch®) is applied to the coating under a specific load and peeled off at a defined angle and speed.<sup>114</sup> The peeling operation is repeated until the coating fails. By analyzing the changes in superhydrophobicity, coating thickness and surface morphology during the peeling test, the mechanical robustness of the coating is evaluated. The core evaluation metric is the total number of peeling cycles required to completely destroy the coating's superhydrophobicity.

### 6.3 Sand impact test

The sand impact test simulates the damage caused by wind-blown sand, dust, or other solid particles impacting the coating



at high velocities. In this test, the coating sample is fixed at a specific angle, and sand particles of a defined size are allowed to fall freely from a fixed height or are accelerated by airflow to strike the coating surface.<sup>147</sup> By analyzing the changes in the coating's superhydrophobicity and surface morphology after impact, the mechanical robustness of the coating is evaluated. The core evaluation metric is the total mass of sand particles required to cause the coating to lose its superhydrophobicity.

In addition to the quantitative evaluation methods mentioned above, several rapid qualitative methods, such as finger abrasion, sharp object scratching, and steel wool abrasion, are also commonly used to assess the mechanical robustness of superhydrophobic coatings.<sup>61,64</sup> Although these methods are not standardized, they provide intuitive and rapid insights into the mechanical robustness of the coating, making them suitable for preliminary screening in the laboratory phase.

## 7. Conclusions and outlook

The strategy of enhancing the mechanical robustness of superhydrophobic coatings with adhesives has garnered significant attention due to its low cost, wide applicability to various substrates, and suitability for large-scale production. However, current coating systems still face several limitations. To further advance the development and practical application of mechanically robust superhydrophobic coatings based on adhesives, it is crucial to comprehensively review the mechanisms, research progress, commercialization status, and common testing methods for the mechanical robustness of these coatings. In this review, we first explored the mechanisms behind adhesive-enhanced superhydrophobic coatings from three perspectives: interfacial adhesion enhancement, structural protection, functional augmentation. We then summarized the progress in mechanically robust superhydrophobic coatings based on different adhesives, highlighting the most used adhesives and identifying the deficiencies in various adhesive systems. Subsequently, we reviewed the progress in mechanically robust superhydrophobic coatings *via* different adhesive application methods, discussing the advantages and disadvantages of each approach. Additionally, we presented an overview of commercially available mechanically robust superhydrophobic coatings based on adhesives, analyzing their mechanical robustness and identifying the best-performing products on the market. Finally, we introduced the common testing methods for the mechanical robustness of superhydrophobic coatings.

Despite the great potential of mechanically robust superhydrophobic coatings based on adhesives and the considerable progress made in basic research, their research and industrial application still face the following challenges.

(1) Limited improvement in mechanical robustness and adverse effects on other properties: superhydrophobicity is primarily determined by the coatings' micro-/nanostructures and low surface energy. However, most adhesives exhibit relatively high surface energy, which, when incorporated into the coatings, often significantly increases the overall surface energy. Furthermore, the introduction of adhesives fills the voids

between low-surface-energy nanoparticles, reducing the roughness of the coatings. These factors collectively degrade the superhydrophobicity of the coatings, particularly the dynamic superhydrophobicity. This remains the largest technical challenge for mechanically robust superhydrophobic coatings based on adhesives. To preserve the micro-/nanostructures and low surface energy, the content of adhesive incorporated into the coating system is often minimal, resulting in only limited improvements in mechanical robustness. Although techniques such as nonsolvent-induced phase separation can mitigate the adverse effects of adhesives on the coating's micro-/nanostructure and chemical composition, the adhesives are still encapsulated by low-surface-energy nanoparticles, which restricts further enhancement of mechanical robustness. Additionally, some adhesives suffer from poor weather resistance, compromising the long-term outdoor durability of superhydrophobic coatings.

(2) Environmental concerns: in the preparation of mechanically robust superhydrophobic coatings based on adhesives, VOCs, such as alcohols, esters, and alkanes, are commonly used as solvents due to their ability to effectively dissolve adhesives. However, the widespread use of VOCs results in the release of substantial amounts of these compounds into the environment during the preparation process, contributing to significant environmental concerns. This poses a major challenge to the practical application of mechanically robust superhydrophobic coatings based on adhesives. Additionally, the use of VOCs complicates the transportation and storage of the coating dispersions, further hindering their commercial viability.

(3) Poor adhesion to hydrophobic substrates: in addition to hydrophilic substrates such as metals, some applications of superhydrophobic coatings involve hydrophobic substrates, such as wind turbine blades coated with fluorocarbon resins or radomes made from materials like polypropylene or ABS. These hydrophobic substrates have low polarity, low surface energy, and lack reactive functional groups, which make it challenging for adhesives to establish effective chemical or physical bonds. As a result, the mechanical robustness of superhydrophobic coatings based on adhesives on these substrates is significantly compromised.

To overcome the challenges encountered by mechanically robust superhydrophobic coatings based on adhesives, we propose several potential research directions.

(1) Efficient screening of commercial adhesives: currently, a wide variety of adhesives are available on the market, making it essential to rapidly identify those most suitable for preparing mechanically robust superhydrophobic coatings. Establishing effective and efficient screening criteria is critical, such as evaluating wettability, mechanical strength, and adhesion to substrates after adhesive curing. Following preliminary screening, selected adhesives can then be combined with low-surface-energy nanoparticles for further optimization.

(2) Synergistic use of commercial adhesives: while each commercial adhesive has its advantages, they also come with limitations, such as inadequate weather resistance for organic adhesives or insufficient hydrophobicity for inorganic adhesives, which can lead to suboptimal overall performance of the



coatings. A promising strategy is to combine two or more adhesives to leverage their respective strengths and offset their weaknesses. However, careful consideration of the compatibility between different adhesives is essential to ensure effective performance.

(3) Development of ideal adhesives: an alternative strategy involves the design and synthesis of ideal block copolymer adhesives that combine excellent adhesion to substrates (including hydrophobic ones), mechanical strength, weather resistance, flexibility, self-healing properties, low surface energy, water solubility, and so on. Although this presents significant challenges, it is a crucial step toward advancing the mechanically robust superhydrophobic coatings based on adhesives.

(4) Combination of nanoparticles of different dimensions: the selection of low-surface-energy nanoparticles is crucial for the development of mechanically robust superhydrophobic coatings based on adhesives. Combining zero-dimensional nanoparticles with one-dimensional nanoparticles (*e.g.*, carbon nanotubes and fibrous clay minerals) is an emerging research direction. The reinforcing effect of one-dimensional nanoparticles can significantly enhance the mechanical robustness of superhydrophobic coatings. Additionally, incorporating nanoparticles of different dimensions can increase the nanoscale roughness of the coatings, thereby effectively improving superhydrophobicity.

(5) Use of functional nanoparticles: the incorporation of functional nanoparticles may significantly enhance the mechanical robustness of superhydrophobic coatings. For example, adding high-hardness, wear-resistant nanoparticles (*e.g.*,  $\text{Al}_2\text{O}_3$  and  $\text{BaSO}_4$ ) can improve the hardness and mechanical strength of the coatings. Additionally, integrating PTFE nanoparticles can reduce the friction coefficient, further boosting the mechanical robustness of superhydrophobic coatings.

(6) Combination of different adhesive application methods: in addition to the choice of materials for superhydrophobic coatings, combining different adhesive application methods can effectively enhance the overall performance of these coatings. For example, when adhesives undergo phase separation, the adhesive becomes encapsulated by low-surface-energy nanoparticles, which reduce adhesion to substrates and limit the mechanical robustness of the coatings. However, by introducing unseparated adhesives or bonding layers, the composite microparticles formed by the phase separated adhesive and low-surface-energy nanoparticles can be firmly anchored to the substrates. This approach could theoretically lead to superhydrophobic coatings that exhibit both excellent superhydrophobicity and mechanical robustness.

(7) Development of waterborne or powder mechanically robust superhydrophobic coatings based on adhesives: environmental concerns represent a significant barrier to the commercialization and practical application of mechanically robust superhydrophobic coatings based on adhesives. Therefore, the development of environmentally friendly alternatives, such as waterborne or powder coatings with excellent overall

performance, is expected to become a leading direction in this field.

(8) Balancing superhydrophobicity, mechanical robustness, and weather resistance: in real-world outdoor environments, superhydrophobic coatings are subjected to factors such as sand and dust erosion, rain impact, UV radiation, and more. Therefore, these coatings must simultaneously exhibit excellent static and dynamic (*i.e.*, impalement resistant) superhydrophobicity, mechanical robustness, weather resistance, *etc.* Achieving a balance between these properties is crucial for ensuring the long-term performance of superhydrophobic coatings in outdoor environments. As such, during the optimization process, it is essential to focus not only on mechanical robustness but also on maintaining high static and dynamic superhydrophobicity, along with weather resistance, to ensure the successful application of superhydrophobic coatings.

## Author contributions

J. F. W. wrote the manuscript. M. Y. M. and B. C. L. contributed to the manuscript preparation. J. P. Z. revised and supervised the manuscript. All authors revised and finalized the manuscript.

## Conflicts of interest

There are no conflicts to declare.

## Data availability

No primary research results, software or code have been included and no new data were generated or analysed as part of this review.

## Abbreviations

APT	Attapulgite
PP	Polypropylene
PS	Polystyrene
BAC	Butyl acetate
EA	Absolute ethanol
FD-POS/PF	Perfluorodecylsilsesquioxane
POS	
SHEP	Self-healing epoxy resin
BAGDE	Bisphenol a diglycidyl ether
PTFE	Polytetrafluoroethylene
PPY	Polypyrrole
HDTMS	Hexadecyltrimethoxysilane
TEOS	Tetraethoxysilane
GO	Graphene oxide
MOCA	4,4'-Methylene bis(2-chloroaniline)
PETMP	Pentaerythritol tetrakis(3-mercaptopropionate)
HFA	1 <i>H</i> ,1 <i>H</i> ,2 <i>H</i> ,2 <i>H</i> -Heptadecafluorodecyl acrylate
FDTP	Fluorinated dithiol prepolymer
PTMEG	Polytetramethylene ether glycol
HMDI	4,4-diisocyanate



PU	Polyurethane
FPU	Fluorinated polyurethane
F-CNPs	Perfluorinated chained nanoparticles

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